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# Nuclear reactions for astrophysics and other applications

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**Abstract.** Cross sections for compound-nuclear reactions are required for many applications. The surrogate nuclear reactions method provides an indirect approach for determining cross sections for reactions on unstable isotopes, which are difficult or impossible to measure otherwise. Current implementations of the method provide useful cross sections for (n,f) reactions, but need to be improved upon for applications to capture reactions.

## 1. Introduction

Compound nuclear reactions play an important role in many applications. Their cross sections are required input for astrophysical models that describe stellar evolution and nucleosynthesis and for modeling processes that are relevant to generating energy. Cross sections for neutron capture, neutron-induced fission, and other processes are required. Often the reactions of interest involve short-lived or highly radioactive target nuclei, which makes it difficult or impossible to measure their cross sections directly. To determine the cross sections of interest, indirect methods, such as inverse-kinematics experiments with radioactive beams, combined with theoretical treatments, are becoming increasingly important. This paper discusses recent progress of the surrogate nuclear reaction method, an indirect approach for determining cross sections for nuclear reactions  $a + A \rightarrow B^* \rightarrow c + C$ , that proceed through the formation of a compound nucleus ( $B^*$ ). The method can be applied when the target is too short-lived to allow for a direct measurement, but a projectile-target combination can be found so that the compound nucleus of interest,  $B^*$ , can be produced in an alternative reaction  $d + D \rightarrow B^* + b$ .

In Sec. 2, representative examples from the fields of nuclear astrophysics and nuclear energy are given to illustrate the importance of cross sections for reactions on unstable targets. In Section 3 an outline of the surrogate approach is given. Some examples of successful applications of the method to (n,f) reactions are shown in Sec. 4. The limitations of presently-employed approximations are illustrated in Sec. 5 for capture reactions.

## 2. Compound reactions for astrophysics and nuclear energy

Cross sections for compound-nuclear reactions are required input for astrophysical models and nuclear-energy simulations.

### *2.1. Nucleosynthesis of heavy elements*

An important goal of nuclear astrophysics research is to explain the origin of the heavy elements ( $A > 56$ ). We have acquired a basic, but incomplete, understanding of the processes that generate

the energy in stars such as our sun, that drive stellar evolution and that are responsible for the synthesis of the elements [1]. Nucleosynthesis of heavy elements beyond  $^{56}\text{Fe}$  is known to take place primarily by neutron capture on lighter seed nuclei in the s (slow neutron-capture) and r (rapid neutron-capture) processes [1, 2, 3], with other processes contributing to the abundances of some specific isotopes. Models of these processes predict relative isotopic abundances for specific astrophysical scenarios. They require nuclear physics input and are constrained by measured isotopic abundances.

The s process proceeds via neutron captures and beta decays through nuclides in and very near the valley of stability. Of particular interest to models of this process are neutron captures on s-process branch points, unstable nuclei with a life time long enough to allow the s process to proceed by either neutron capture or  $\beta$  decay. The strength with which one path dominates over the other depends on environmental variables, such as neutron density, temperature, and pressure, as well as on nuclear properties, specifically capture rates and beta-decay life times. Information on the astrophysical conditions can be inferred if the nuclear properties are known.

Beyond being important for gaining a more detailed description of the s process environments, reliable s-process abundances are crucial for our understanding of the r (rapid neutron-capture) process. The r process takes place in an environment with high temperature and high neutron flux. In such conditions the average time between neutron captures is much shorter than the life time for  $\beta$ -decay and reaction flows can proceed to very neutron-rich nuclei. Open questions include the exact path along the nuclear chart of the neutron captures and beta decays involved and the astrophysical site(s) of the process. Typically, r-process abundances are inferred by subtracting calculated s-process abundances from measured total abundances. This requires detailed calculations to predict s-process abundances, and reliable cross sections for neutron captures along the s-process path. Beta-decay half lives and nuclear masses, which determine nucleon separation energies, are considered to be the most important nuclear physics inputs for r-process models, as they determine the neutron capture path in scenarios that assume  $(n, \gamma)$  and  $(\gamma, n)$  reactions to be in equilibrium with each other, but models that do not invoke the equilibrium assumption require neutron capture rates for thousands of unstable nuclei.

## 2.2. Nuclear energy

The world-wide growing demand for energy, coupled with increasing concern over pollution resulting from burning of fossil fuels, has resulted in renewed interest in nuclear energy. To exploit nuclear energy in a clean, efficient, and safe manner, concerns regarding reactor safety, waste handling, proliferation risks, and economic competitiveness have to be addressed.

Advanced nuclear-energy systems may recycle actinides produced in conventional reactors or take advantage of alternative fuel cycles. A thorium-based fuel cycle, for instance, is appealing, since it produces less radiotoxic heavy transuranium isotopes than the conventional uranium-plutonium fuel cycle; thorium is also more abundant in the earth's crust than uranium. An important component of the research and development for these reactor concepts is the improvement of the fundamental nuclear data [4]. The transuranic nuclides, for example, play a much more prominent role in these new designs and yet the available cross-section data is quite limited. In addition, neutron-capture reactions could be used to incinerate long-lived fission fragments and therefore limit the challenges associated with the reactor waste.

Sensitivity studies [5] indicate that high-quality, reliable cross-section data are needed for neutron-induced reactions for a wide variety of radioactive isotopes covering neutron energies from thermal up to tens of MeV. In particular, capture and fission reactions on many of the isotopes of thorium, uranium, plutonium, and the minor actinides (such as  $^{237}\text{Np}$ ,  $^{241-243}\text{Am}$ , and  $^{244-245}\text{Cm}$ ), as well as certain long-lived fission fragments. Many of these cross sections are extremely challenging to measure directly but significantly contribute to uncertainties in the reliable design and safe operation of a nuclear-energy system.

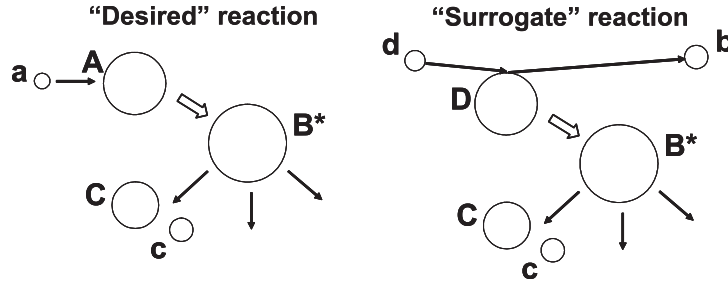
Some relevant cross sections, such as the  $(n, f)$  cross sections for  $^{241}\text{Am}$ ,  $^{243-244}\text{Cm}$  have already been determined with the surrogate approach [6, 7, 8, 9, 10]. It is anticipated that the surrogate reaction method will continue to provide valuable cross-section results on isotopes for which there is limited, poor-quality, or no data of use for nuclear-energy applications.

### 3. The surrogate idea

The appropriate formalism for the description of a compound-nuclear reaction  $a + A \rightarrow B^* \rightarrow c + C$  is a statistical one [11]. Based on Bohr’s hypothesis of the independence of formation and decay of the compound nucleus (CN), the cross sections are calculated in the framework of the Hauser-Feshbach formalism, which properly takes account of the conservation of angular momentum and parity in the reaction:

$$\sigma_{\alpha\chi}(E_a) = \sum_{J,\pi} \sigma_{\alpha}^{CN}(E_{ex}, J, \pi) G_{\chi}^{CN}(E_{ex}, J, \pi), \quad (1)$$

with  $\alpha$  and  $\chi$  denoting the relevant entrance and exit channels,  $a + A$  and  $c + C$ , respectively. The excitation energy  $E_{ex}$  of the compound nucleus,  $B^*$ , is related to the center-of-mass energy  $E_a$  in the entrance channel via the energy needed for separating  $a$  from  $B$ :  $E_a = E - S_a(B)$ . In many cases the formation cross section  $\sigma_{\alpha}^{CN} = \sigma(a + A \rightarrow B^*)$  can be calculated to a reasonable accuracy by using optical potentials, while the theoretical decay probabilities  $G_{\chi}^{CN}$  for the different decay channels  $\chi$  are often quite uncertain. The latter are difficult to calculate accurately since they require knowledge of optical models, level densities, and strength functions for the various possible exit channels. The objective of the surrogate method is to determine or constrain these decay probabilities experimentally.



**Figure 1.** Schematic representation of the “desired” (left) and “surrogate” (right) reaction mechanisms. The basic idea of the surrogate approach is to replace the first step of the desired reaction,  $a + A$ , by an alternative (surrogate) reaction,  $d + D \rightarrow b + B^*$ , that populates the same compound nucleus. The subsequent decay of the compound nucleus into the relevant channel,  $c + C$ , can then be measured and used to extract the desired cross section.

In the surrogate approach, the compound nucleus  $B^*$  is produced by means of an alternative (“surrogate”), direct reaction,  $d + D \rightarrow b + B^*$ , and the desired decay channel  $\chi(B^* \rightarrow c + C)$  is observed in coincidence with the outgoing particle  $b$  (see Fig. 1). The probability for forming  $B^*$  in the surrogate reaction (with specific values for  $E_{ex}$ ,  $J$ ,  $\pi$ ) is  $F_{\delta}^{CN}(E_{ex}, J, \pi)$ , where  $\delta$  refers to the entrance channel reaction  $D(d, b)$ . The quantity

$$P_{\delta\chi}(E_{ex}) = \sum_{J,\pi} F_{\delta}^{CN}(E_{ex}, J, \pi) G_{\chi}^{CN}(E_{ex}, J, \pi), \quad (2)$$

which gives the probability that the compound nucleus  $B^*$  was formed with energy  $E_{ex}$  and decayed into channel  $\chi$ , can be obtained experimentally, by measuring  $N_{\delta}$ , the total number

of surrogate events, and  $N_{\delta\chi}$ , the number of coincidences between the direct-reaction particle and the observable that identifies the relevant exit channel:  $P_{\delta\chi}^{exp}(E_{ex}) = N_{\delta\chi}/N_{\delta\epsilon\delta}$ . Here,  $\epsilon_{\delta}$  denotes the efficiency for detecting the outgoing direct-reaction particle  $b$  and the exit channel  $\chi$ . To simplify the notation, we suppress the dependence of the coincidence probability  $P_{\delta\chi}(E_{ex})$  on the angle  $\theta_b$  of the outgoing direct-reaction particle  $b$ .

The distribution  $F_{\delta}^{CN}(E_{ex}, J, \pi)$ , which may be very different from the CN spin-parity populations following the absorption of the projectile  $a$  in the desired reaction, has to be determined theoretically, so that the branching ratios  $G_{\chi}^{CN}(E_{ex}, J, \pi)$  can be extracted from the measurements. In practice, the decay of the CN is modeled and the  $G_{\chi}^{CN}(E_{ex}, J, \pi)$  are obtained by adjusting parameters in the model to reproduce the measured probabilities  $P_{\delta\chi}(E_{ex})$  [12, 13]. Subsequently, the sought-after cross section can be obtained by combining the calculated cross section  $\sigma_{\alpha}^{CN}(E_{ex}, J, \pi)$  for the formation of  $B^*$  (from  $a+A$ ) with the extracted decay probabilities  $G_{\chi}^{CN}(E_{ex}, J, \pi)$  for this state, see Eq. 1.

*Weisskopf-Ewing Approximation.* Under certain circumstances, the decay of the intermediate equilibrated system, the compound nucleus, becomes independent of its angular momentum and parity, and the cross section for the reaction factorizes into a simple product of a formation cross section and a decay probability for the exit channel of interest [11]:

$$\sigma_{\alpha\chi}(E_a) = \sigma_{\alpha}^{CN}(E_{ex}) \mathcal{G}_{\chi}^{CN}(E_{ex}), \quad (3)$$

where  $\sigma_{\alpha}^{CN}(E_{ex}) = \sum_{J,\pi} \sigma_{\alpha}^{CN}(E_{ex}, J, \pi)$  is the reaction cross section describing the formation of the compound nucleus in the desired reaction and  $\mathcal{G}_{\chi}^{CN}(E_{ex})$  denotes the  $J^{\pi}$ -independent decay probability for the exit channel  $\chi$ . This is the Weisskopf-Ewing (WE) limit of the Hauser-Feshbach theory [11]. In the context of surrogate reactions, the Weisskopf-Ewing approximation greatly simplifies the application of the method: It becomes straightforward to obtain the  $J^{\pi}$ -independent branching ratios  $\mathcal{G}_{\chi}^{CN}(E_{ex})$  from measurements of  $P_{\delta\chi}(E_{ex}) [= \mathcal{G}_{\chi}^{CN}(E_{ex})$  since  $\sum_{J,\pi} F_{\delta}^{CN}(E_{ex}, J, \pi) = 1]$  and to calculate the desired reaction cross section. Calculating the direct-reaction probabilities  $F_{\delta}^{CN}(E_{ex}, J, \pi)$  and modeling the decay of the compound nucleus are no longer required. Most applications to date invoke the Weisskopf-Ewing approximation.

#### 4. Neutron-induced fission reactions

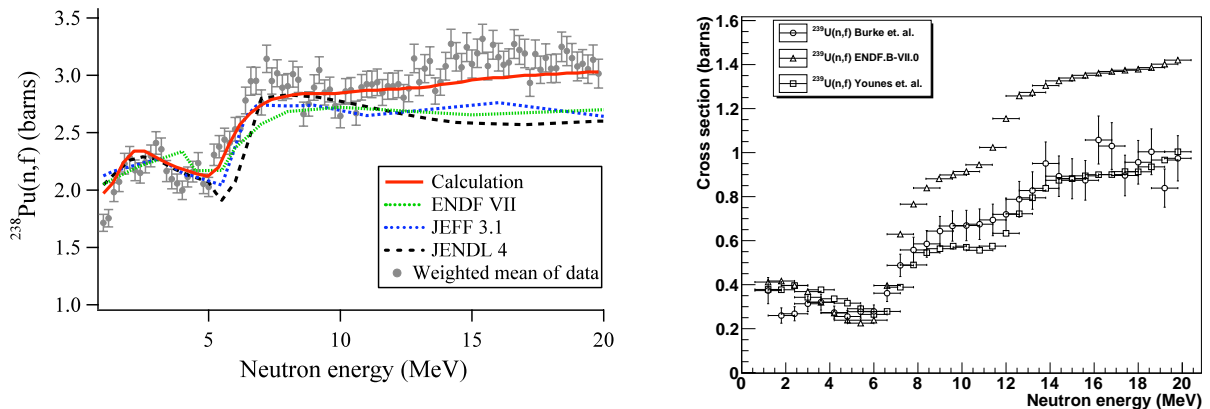
The surrogate approach was first employed in the 1970s to estimate neutron-induced fission cross sections from transfer reactions. These early applications of the method made use of the Weisskopf-Ewing approximation: The transfer reactions of the 1970s [14, 15] produced (n,f) cross section estimates for various actinide targets which agreed with direct measurements (where available) to about 10-20% for incident neutron energies above 1 MeV. Discrepancies at lower energies were later attributed to large uncertainties in the low-energy optical model employed, and the use of the Weisskopf-Ewing approximation [12, 13].

More recently, a French group has carried out surrogate experiments at the Institut de Physique Nucléaire (IPN) in Orsay to determine cross sections for neutron-induced reactions on several minor actinide nuclei relevant to the thorium-uranium fuel cycle and the transmutation of nuclear waste [6, 7, 10]. The transfer reactions  $^{232}\text{Th}(^3\text{He},x)$  and  $^{243}\text{Am}(^3\text{He},x)$ , with  $x=\alpha, t, d, p$ , were employed to obtain (n,f) and (n, $\gamma$ ) cross sections for Th and Pa targets [6, 7] and (n,f) cross sections for Cm and Am targets [10], respectively. The analyses assumed that the Weisskopf-Ewing approximation is valid. The extracted (n,f) cross sections were found to be consistent with known directly-measured cross section measurements, where these were available. They proved useful for resolving controversies between discrepant measurements and for providing data for previously unavailable energy regions.

The STARS/LiBerACE collaboration in the United States has carried out a number of experiments at the 88-inch cyclotron at Lawrence Berkeley National Laboratory. Light-ion

beams have been used for inelastic scattering, charge exchange, and one- or two-neutron transfer reactions. Fission cross sections, e.g. the  $^{237}\text{Np}(n,f)$  cross section [16], have been determined using the Weisskopf-Ewing approximation or a variant thereof, the surrogate ratio method.

The *Surrogate Ratio approach* [17, 18, 19] requires the (approximate) validity of the Weisskopf-Ewing limit. The ratio  $R(E) = \sigma_{\alpha_1\chi_1}/\sigma_{\alpha_2\chi_2}$  of the cross sections of two CN reactions is measured, using two surrogate experiments. An independent determination of the cross section  $\sigma_{\alpha_2\chi_2}$  can then be used to deduce  $\sigma_{\alpha_1\chi_1}$ . An advantage of using the ratio method is the fact that it eliminates the need to accurately measure the total number of surrogate reaction events ( $N_\delta$ ), since one determines the ratio of coincidence probabilities  $P_{\delta_1\chi_1}/P_{\delta_2\chi_2}$ , rather than an absolute probability  $P_{\delta\chi} = N_{\delta\chi}/N_\delta$ . Furthermore, there are indications that small to moderate deviations from the WE limit cancel in this approach [17]. Cross sections for (n,f) reactions extracted in the ratio approximation have been tested for consistency with results from direct measurements, complementary surrogate experiments [19, 20], and theoretical simulations [17]. For (n,f) reactions one typically finds that the spin-parity mismatch between the desired and surrogate reactions has a much smaller effect on the extracted cross section than in an approach that uses the WE approximation and absolute probabilities. Also, deviations caused by pre-equilibrium effects are diminished, thus improving the overall agreement between extracted and expected cross sections.



**Figure 2.** Fission cross sections obtained from surrogate ratio measurements. The  $^{238}\text{Pu}(n,f)$  cross section (left panel) was determined relative to the known  $^{234}\text{U}(n,f)$  and  $^{235}\text{U}(n,f)$  cross sections [24]. The weighted average of the two measurements is compared to several evaluations; the solid line represents a calculation based on the new data. The  $^{239}\text{U}(n,f)$  cross section (right panel) was determined relative to the known  $^{235}\text{U}(n,f)$  cross section [21]; here it is compared to the result of an earlier surrogate measurement [22] and to the ENDF/B-VII evaluation [23].

Two recent applications of the ratio approach are shown in Figure 2. The  $^{238}\text{Pu}(n,f)$  cross section, which is needed for reactor applications and for transmutation studies, was recently determined using inelastic scattering surrogate reactions [24]. Ressler *et al* [24] produced the compound nuclei  $^{239}\text{Pu}^*$ ,  $^{235}\text{U}^*$ , and  $^{236}\text{U}^*$  via inelastic  $\alpha$  scattering. Surrogate ratio analyses yielded the desired  $^{238}\text{Pu}(n,f)$  cross section relative to both the  $^{234}\text{U}(n,f)$  and the  $^{235}\text{U}(n,f)$  cross sections. Since the latter two are known, the  $^{238}\text{Pu}(n,f)$  cross section could be extracted; the weighted average of both measurements is shown in the left panel of Figure 2. The two measurements are in good agreement with each other for 5-20 MeV and the averaged cross section agrees well with previous, direct, measurements in the 5-10 MeV range; it is somewhat higher (by less than 20%) than those at about 15 MeV. The ratio measurement does not, and

is not expected to, produce highly-accurate fission results at low energies ( $< 5$  MeV here), due to the underlying Weisskopf-Ewing assumption. It does, however, provide continuous data for neutron energies from 5 to 20 MeV, and supplement earlier measurements, which were sparse in the energy regime around 10-15 MeV.

The right panel of Figure 2 shows the  $^{239}\text{U}(\text{n},\text{f})$  cross section (open circles), which was recently determined relative to the known  $^{235}\text{U}(\text{n},\text{f})$  cross section [21]. The CN  $^{240}\text{U}^*$  and  $^{236}\text{U}^*$  were produced via the two-neutron transfer reactions  $^{238}\text{U}(^{18}\text{O},^{16}\text{O})$  and  $^{234}\text{U}(^{18}\text{O},^{16}\text{O})$ , respectively, and  $^{16}\text{O}$  – fission fragment coincidence events were counted. The extracted  $^{239}\text{U}(\text{n},\text{f})$  cross section agrees well with the result of an earlier surrogate experiment (open squares, from Ref. [22]); both are lower than the ENDF/B-VII evaluation (open triangles). No direct measurements exist, since the half life of  $^{239}\text{U}$  is only 23.4 minutes.

The literature and the above examples indicate that (n,f) cross sections extracted from surrogate data are typically consistent with direct measurements (where available) and/or other surrogate measurements, despite the approximations used. In addition, calculations, which test the approximation schemes employed in the analyses of surrogate fission data [17], illustrate the level of accuracy that one can under reasonable circumstances expect from the surrogate approach. Discrepancies between indirectly and directly measured cross sections are often less than 10%. The largest deviations are found at low energies, where the Weisskopf-Ewing approximation is not expected to be valid; in those cases, it becomes necessary to account for the differences in the spin-parity distributions occurring in the desired and surrogate reactions.

## 5. Capture reactions

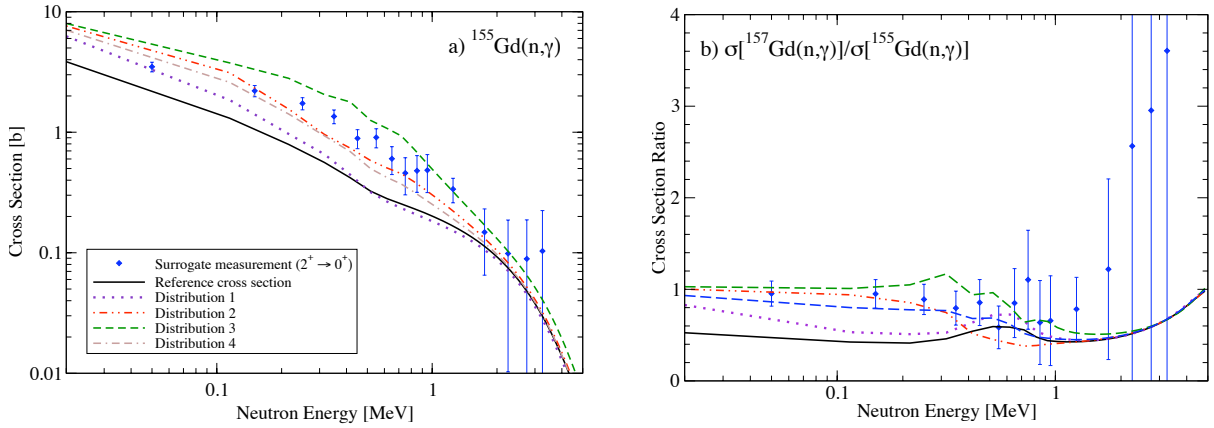
Capture cross sections provide specific challenges for the surrogate approach. First, the level of precision required for the cross section is often higher than in the fission case: Recent advances in modeling the astrophysical s process have resulted in requests to determine capture cross sections within a few percent and nuclear-energy applications require cross sections to within 5-10% [25, 5]. Achieving an accuracy of a few percent is challenging, but constraining an unknown (n, $\gamma$ ) cross section to within 20-30% should be considered a meaningful improvement of the situation, in particular since current cross section evaluations often show large deviations from each other. Secondly, it is the low-energy regime that is relevant to many applications. For s-process applications, for example, one needs cross sections from a few keV to about 200 keV. Both calculations and measurements have shown that this is the energy range for which the Weisskopf-Ewing approximation typically breaks down.

Theoretical studies have been carried out to assess the feasibility of obtaining capture cross sections from surrogate measurements and to determine promising candidates for such measurements. The strategy followed in these investigations is to extract information from Hauser-Feshbach calculations that have been adjusted to reproduce known cross sections (capture and, where applicable, fission). The branching ratios (channel probabilities),  $G_{\chi}^{CN}(E, J, \pi)$  that enter Eqs. 1 and 2 can be calculated via this procedure; the Weisskopf-Ewing limit is reached when the branching ratios are approximately equal.

Overall, the studies show that the probability for a compound nucleus to decay via  $\gamma$  emission ( $\chi = \gamma$ ) depends sensitively on the spin-parity population of the nucleus prior to decay. The dependence of the  $\gamma$ -branching ratios on the  $J\pi$  distribution is greater than that found previously for fission ( $\chi = \text{fission}$ ). Calculations for representative Zirconium, Gadolinium, and Uranium nuclei showed a strong dependence of the  $\gamma$  branching ratios on the spins populated in the compound nucleus. The effect was particularly strong for the  $^{92}\text{Zr}$  nucleus, which has a closed proton subshell ( $Z=40$ ) and a nearly-closed neutron shell ( $N=52 \approx 50$ ) [26]. A comparison with the results for Gadolinium and Uranium confirms the notion that the higher level densities present in the deformed rare-earth and actinide regions do reduce the sensitivity of the  $\gamma$ -decay probabilities to compound-nuclear spin-parity distributions and nuclear-structure effects [27].



For Gadolinium, we demonstrate in Fig. 3a that the  $(n,\gamma)$  cross sections obtained from a Weisskopf-Ewing analysis of surrogate data can differ significantly from the expected ‘true’ cross section. Shown are the results of a Weisskopf-Ewing analysis of the  $^{156}\text{Gd}(p,p'\gamma)$  coincidence data measured by Scielzo *et al* [28] (symbols with error bars) and theory results of a sensitivity study by Escher and Dietrich [27] (dashed and dotted curves). The  $^{155}\text{Gd}(n,\gamma)$  cross section extracted from the indirect measurement is a factor of 2-3 larger than the directly-measured cross section (solid curve). It falls, for the most part, between the theoretical curves. The latter were obtained by combining calculated branching ratios and schematic spin-parity distributions to simulate surrogate measurements,  $P_{\delta\gamma}^{sim}(E) = \sum_{J,\pi} F_{\delta}^{CN}(E, J, \pi) G_{\gamma}^{CN}(E, J, \pi)$ , and carrying out a WE ‘analysis’ to obtain the desired cross section,  $\sigma_{n,\gamma}^{WE,sim}(E) = \sigma_n^{CN}(E) P_{\delta\gamma}^{sim}(E)$ , where  $\sigma_n^{CN}(E)$  denotes the CN formation cross section. The range of cross sections,  $\sigma_{n,\gamma}^{WE,sim}(E)$ , obtained by varying the simulated spin distributions within reasonable limits provides a measure of the uncertainty in the extracted cross section due to the use of the WE approximation.



**Figure 3.** a) Weisskopf-Ewing estimate for the  $^{155}\text{Gd}(n,\gamma)$  cross section. The cross section extracted from a surrogate  $^{156}\text{Gd}(p,p'\gamma)$  measurement [28] is compared to WE ‘analyses’ of simulated surrogate experiments [27], for different CN spin-parity distributions, and to the reference cross section, which is a fit to directly-measured cross sections. b) Cross section ratio obtained from (measured and simulated) surrogate data, compared to the ratio of evaluated cross sections. The spin-parity distributions used in the simulations are from Ref. [27], Fig. 10.

The studies of the Gadolinium region also show that the WE approximation overestimates the  $(n,\gamma)$  cross section by factors which depend on the nucleus under consideration [27, 28]. For the  $^{155}\text{Gd}(n,\gamma)$  and  $^{157}\text{Gd}(n,\gamma)$  cross sections, the differences are large, despite the structural similarities of the relevant nuclei. Consequently, the ratio approach results in  $^{155}\text{Gd}(n,\gamma)$  cross sections that are too large by up to a factor two for energies below about  $E_n = 0.7$  MeV (Fig. 3b). The effect is seen in both the theoretical sensitivity study and the experimental results. Overall, the ratio approach was found to reduce, but not eliminate, the effect of the spin-parity mismatch on the extracted cross sections for energies where the WE assumption is a poor approximation.

## 6. Concluding Remarks

Indirect approaches have to be developed in order to provide much-needed nuclear data, in particular cross sections for reactions on unstable isotopes. We have reviewed recent application of the surrogate nuclear reaction method, which aims at providing cross section information for compound nuclear reactions. Past applications of the surrogate method have demonstrated that it can provide useful cross section estimates for neutron-induced fission of actinides. Similar success in applications to neutron capture for a range of isotopes would be very valuable and

remains to be demonstrated. Most analyses of the fission data carried out so far have made approximations that are likely to break down in situations relevant for extracting  $(n,\gamma)$  cross sections from surrogate measurements, making this a more challenging reaction to tackle. The examples discussed show this breakdown for capture reactions on Gadolinium isotopes. They underscore the need to take into account the different spin-parity distributions that occur in the desired and surrogate reactions. Work to address this issue is underway [29]

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